

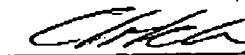
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Acting Managing Director of RWS Group Ltd UK Translation Division, of Europa House,  
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1. That I am a citizen of the United Kingdom of Great Britain and Northern Ireland.
2. That the translator responsible for the attached translation is well acquainted with the German and English languages.
3. That the attached is, to the best of RWS Group Ltd knowledge and belief, a true translation into the English language of the accompanying copy of the specification filed with the application for a patent in Germany on 3 March 2003 under the number 103 09 385.0 and the official certificate attached hereto.
4. That I believe that all statements made herein of my own knowledge are true and that all statements made on information and belief are true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the patent application in the United States of America or any patent issuing thereon.



For and on behalf of RWS Group Ltd

The 9th day of October 2006

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**Priority Certificate**  
**DE 103 09 385.0**  
**for the filing of a Patent Application**

**File Reference:** 103 09 385.0

**Filing date:** 03 March 2003

**Applicant/Proprietor:** Clariant Produkte (Deutschland) GmbH,  
65929 Frankfurt/DE

(formerly: Clariant GmbH)

**Title:** Flame retardant and stabilizer combined for thermoplastic polymers

**IPC:** C 08 K, C 08 L

The attached documents are a correct and accurate reproduction of the original submission for this Application.

Munich, 20 September 2006  
German Patent and Trademark Office

The President

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Kahle

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Dr. TP

## Description

## 5 Flame retardant and stabilizer combined for thermoplastic polymers

The invention relates to a flame retardant and stabilizer combined for thermoplastic polymers, and to polymeric molding compositions which comprise these flame retardants and stabilizers combined.

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With a few exceptions, the processing of thermoplastics takes place in the melt. The attendant changes in structure and conditions bring about alterations of chemical structure in almost all plastics. The consequence can be crosslinking, oxidation, molecular-weight changes, and also resultant changes in physical and technical

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properties. In order to reduce the adverse effect on the polymers during processing, various additives are added, depending on the plastic. Stabilizers are generally added, and these suppress, or at least retard, the alteration processes, such as reactions involving crosslinking or involving degradation. Lubricants are also admixed with most plastics, the main function of these being to improve the flow

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behavior of the melt.

A wide variety of different additives is generally used simultaneously, each of these assuming one function. For example, antioxidants and stabilizers are used in order that the plastic withstand processing with no chemical degradation, and then be

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stable over a long period with respect to external effects, such as heat, UV light, weathering and oxygen (air). Lubricants not only improve flow behavior but also inhibit excessive adhesion of the plastics melt to hot machinery parts, and act as dispersing agents for pigments, fillers, and reinforcing materials.

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The use of flame retardants can have an effect on the stability of the plastic during processing in the melt. Large amounts of flame retardants often have to be added in order to give the plastic adequate flame retardancy complying with international standards. The chemical reactivity of flame retardants, which is required for flame-

retardant action at high temperatures, can result in impairment of the processing stability of plastics. By way of example, the result can be increased polymer degradation, crosslinking reactions, evolution of gases, or discoloration. When plastics are processed without flame retardant, some of these effects do not occur at all, or occur only to a reduced extent.

When no flame retardants are added, polyamides are generally stabilized by small amounts of copper halides, or else by aromatic amines and sterically hindered phenols, the main aim being to achieve long-term stability at high long-term service temperatures: "Plastics Additives Handbook", 5<sup>th</sup> Edition, (H. Zweifel (ed.), Carl Hanser Verlag, Munich, 2000, pages 80 to 84).

Particularly for thermoplastic polymers, the salts of phosphinic acids (phosphinates) have proven to be effective flame-retardant additives (DE-A-2 252 258 uad DE-A-2 447 727). Calcium phosphinates and aluminum phosphinates have been described as having particularly effective action in polyesters, and impair the properties of the polymer molding composition materials to a lesser degree than, by way of example, the alkali metal salts (EP-A-0 699 708).

Synergistic combinations of phosphinates with certain nitrogen-containing compounds have also been found, and these have more effective action as flame retardant than the phosphinates alone in very many polymers (PCT/EP97/01664, and also DE-A-197 34 437 and DE-A-197 37 727).

Carbodiimides, isocyanates, and isocyanurates have proven effective for stabilizing polymer molding compositions with phosphorus-containing flame retardants (DE-A-199 20 276).

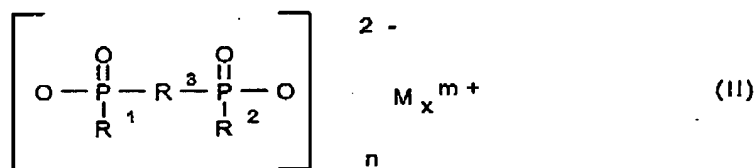
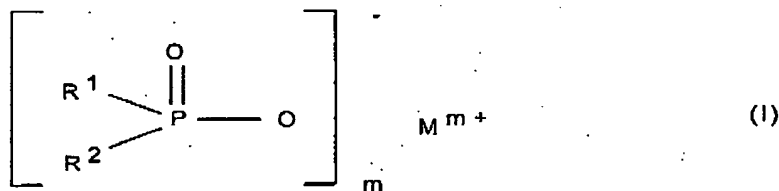
In particular when phosphorus-containing flame retardants are used in polyamides, the action of the stabilizers described hitherto has proven to be insufficient, especially for suppressing effects arising during processing, such as discoloration and molecular-weight degradation.

DE-A-196 14 424 describes phosphinates in combination with nitrogen synergists in polyesters and in polyamides. DE-A-199 33 901 describes phosphinates in combination with melamine polyphosphate as flame retardants for polyesters and polyamides. However, when these newly developed, highly active flame retardants are used the result can be partial polymer degradation, and also discoloration of the polymer, in particular at processing temperatures above 300°C.

It was therefore an object of the present invention to provide flame retardant combinations for thermoplastics, where these provide not only flame retardancy but also a stabilizing action with respect to the plastic. This object is achieved by adding basic or amphoteric oxides, hydroxides, carbonates, silicates, borates, stannates, mixed oxide/hydroxides, oxide/hydroxide/carbonates, hydroxide/silicates or hydroxide/borates, or a mixture of these substances (component C) in combination with a mixture composed of a phosphonite or a phosphonite/phosphite mixture (component D) and an ester or salt of montan wax acid (component E), or a combination composed of components D or E with an aromatic di- or tricarboxylic ester or aromatic di- or tricarboxamide (component F), or with a combination of D, E, and F, when phosphinates (component A) or a mixture of these are used with nitrogen-containing synergists (component B) as flame retardants.

The invention therefore provides a flame retardant and stabilizer combined, for thermoplastic polymers, which comprises, as component A, from 25.0 to 99.8% by weight of a phosphinic salt of the formula (I) and/or a diphosphinic salt of the formula (II) and/or polymers of these,

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where

$\text{R}^1, \text{R}^2$  are identical or different and are  $\text{C}_1\text{-C}_6$ -alkyl, linear or branched, and/or aryl;

5  $\text{R}^3$  is  $\text{C}_1\text{-C}_{10}$ -alkylene, linear or branched,  $\text{C}_6\text{-C}_{10}$ -arylene, -alkylarylene or -arylalkylene;

$\text{M}$  is Mg, Ca, Al, Sb, Sn, Ge, Ti, Zn, Fe, Zr, Ce, Bi, Sr, Mn, Li, Na, K, and/or a protonated nitrogen base;

$m$  is 1 to 4;

10  $n$  is 1 to 4;

$x$  is 1 to 4;

and comprises, as component B,

from 0 to 75% by weight of a nitrogen-containing synergist or of a

phosphorus/nitrogen flame retardant, and comprises, as component C, from 0.1 to

15 50% by weight of a basic or amphoteric oxide, hydroxide, carbonate, silicate, borate,

stannate, mixed oxide/hydroxide, oxide/hydroxide/carbonate, hydroxide/silicate, or

hydroxide/borate, or a mixture of these substances, and comprises, as component

D, from 0 to 5% by weight of a mixture composed of a phosphonite or of a

phosphonite/phosphite mixture, and comprises, as component E, from 0 to 5% by

20 weight of an ester or salt of montan wax acid, and comprises, as component F, from

0.1 to 5% by weight of an aromatic di- or tricarboxylic ester or aromatic di- or

tricarboxamide, the entirety of the components always being 100% by weight.

Surprisingly, it has been found that inventive combinations of phosphinates and, where appropriate, nitrogen synergists, such as melamine polyphosphate, exhibit markedly improved stability on incorporation into polymers if certain oxides, hydroxides, carbonates, silicates, borates, stannates, mixed oxide/hydroxides, oxide/hydroxide/carbonates, hydroxide/silicates, or hydroxide/borates, or a mixture of these substances, in combination with mixtures composed of a phosphonite or of a phosphonite/phosphite mixture, and an ester or salt of montan wax acid, or a combination of these components with an aromatic di- or tricarboxylic ester or aromatic di- or tricarboxamide, or a combination of these are added. The specified oxides, hydroxides, carbonates, silicates, borates, stannates, mixed oxide/hydroxides, oxide/hydroxide/carbonates, hydroxide/silicates, or hydroxide/borates, or a mixture of these substances can suppress discoloration, but polymer degradation is observed. Surprisingly, it has been found that polymer degradation can also be inhibited by adding certain other additives.

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The inventive combinations reduce discoloration of the plastics during processing in the melt, and suppress the degradation of the plastics to give units with lower molecular weight, together with full retention of flame retardancy.

20 M is preferably calcium, aluminum or zinc.

The protonated nitrogen bases are preferably the protonated bases of ammonia, melamine, triethanolamine, in particular  $\text{NH}_4^+$ .

25  $\text{R}^1$  and  $\text{R}^2$ , identical or different, are preferably  $\text{C}_1$ - $\text{C}_6$ -alkyl, linear or branched, and/or phenyl.

$\text{R}^1$  and  $\text{R}^2$ , identical or different, are particularly preferably methyl, ethyl, n-propyl, isopropyl, n-butyl, tert-butyl, n-pentyl and/or phenyl.

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$\text{R}^3$  is preferably methylene, ethylene, n-propylene, isopropylene, n-butylene, tert-butylene, n-pentylene, n-octylene, or n-dodecylene.

Another preferred meaning of  $\text{R}^3$  is phenylene or naphthylene.

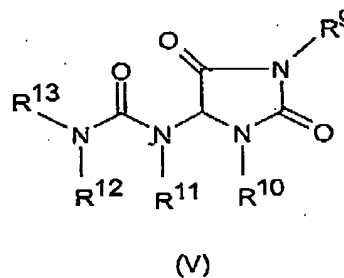
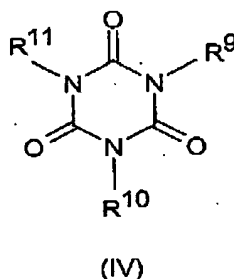
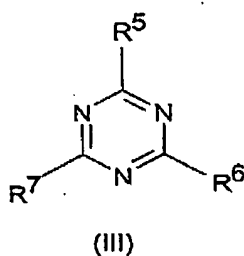
Suitable phosphinates have been described in PCT/WO97/39053, expressly incorporated herein by way of reference.

- 5 Particularly preferred phosphinates are aluminum phosphinates, calcium phosphinates, and zinc phosphinates.

- 10 Synergistic combinations of the specified phosphinates with nitrogen-containing compounds which have more effective action as flame retardants than the phosphinates alone in very many polymers (DE-A-196 14 424, DE-A-197 34 437, and DE-A-197 37 727) are also in accordance with the invention. The flame-retardant action of the phosphinates may be improved via combination with other flame retardants, preferably nitrogen-containing synergists, or phosphorus/nitrogen flame retardants.

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The nitrogen-containing synergists are preferably those of the formulae (III) to (VIII), or a mixture of these



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